Nuclear hexadecapole antishielding factors

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Using Hartree-Fock-Slater wave functions, values of the nuclear hexadecapole antishielding factor η_∞ have been calculated for the following ions: Ti^{3+}, Mn^{2+}, Cu^{+}, Zn^{2+}, Ge^{4+}, Rb^{+}, Zr^{3+}, Nb^{5+}, Tc^{2+}, Ag^{+}, In^{3+}, Sb^{3+}, Cs^{+}, Hf^{3+}, Re^{2+}, Au^{+}, Hg^{2+}, Bi^{3+}, Fr^{+}, Th^{3+}, and Am^{2+}. The radial distortions induced in the core electrons due to nuclear hexadecapole moment were calculated using Sternheimer's perturbation numerical procedure. The contribution to η_∞ due to the *nf* shell is much weaker in magnitude than that from the *nd* shell. In the three *nd* series of ions considered here, maximum values of η_∞ are obtained in the cases of Cu^{+}, Ag^{+}, and Au^{+} ions, respectively, where the outer *d* shell is complete and is most external. With the assumption that a large nuclear quadrupole moment may be associated with a significant hexadecapole moment, it is suggested that of the systems examined here containing nuclei with spin $I \geq 2$, Bi^{3+}, Re^{2+}, Sb^{3+}, In^{3+}, and Mn^{2+} are likely to exhibit prominently nuclear hexadecapole interaction effects since these have favorable natural abundance, quadrupole moment, magnetic resonance sensitivity, and large η_∞ values.

I. INTRODUCTION

Nuclei with spin $I \geqslant 2$ can possess a nonvanishing electric hexadecapole moment H which interacts with the triple gradient of the surrounding charges giving rise to a nuclear electric hexadecapole interaction. In 1955, Wang[1] inferred that an unexplained shift in ^{121}Sb ($I=\frac{5}{2}$) and ^{123}Sb ($I=\frac{7}{2}$) nuclear quadrupole energy levels was due to the static nuclear electric hexadecapole interaction. In 1966, Mahler, James, and Tantilla[2] observed the externally induced hexadecapole transition between magnetically split ^{115}In ($I=\frac{9}{2}$) levels in InAs. Dinesh and Smith[3] have reported observation of nuclear hexadecapole interaction from their nuclear quadrupole resonance (NQR) study of ^{93}Nb ($I=\frac{9}{2}$) in $NbCl_5$. Recently, Dankwort, Ferch, and Gebauer[4] have detected a nuclear hexadecapole interaction in the ground state of ^{165}Ho ($I=\frac{7}{2}$) using the atomic-beam magnetic-resonance method. The effects of the nuclear hexadecapole interaction are masked as a result of the presence of much stronger nuclear quadrupole interaction, and hence are more difficult to detect experimentally. Analogous to the nuclear quadrupole antishielding factor[5] γ_∞, it was shown by Sternheimer[6] that the hexadecapole moments induced in the electronic closed shells (d and f) give rise to a large antishielding (enhancement) of the nuclear hexadecapole moment H which might facilitate the experimental detection of the hexadecapole interaction in favorable cases.

The hexadecapole moment induced in the electronic closed shell H_{ind} can be written

$$H_{\text{ind}} = -\eta_\infty H, \tag{1}$$

where η_∞ is the hexadecapole antishielding factor. The total nuclear hexadecapole moment for the ion now becomes

$$H_{\text{ion}} = H + H_{\text{ind}} = (1-\eta_\infty)H. \tag{2}$$

So far, η_∞ values have been calculated only for five ions. Thus, using Hartree-Fock (HF) wave functions[7-9] for Cu^{+}, Ag^{+}, and Cs^{+}, Hartree wave functions[10] for Hg^{2+}, and Hartree-Fock-Slater (HFS) wave functions[11] for In, Sternheimer[12-14] earlier reported the following values of η_∞: $\eta_\infty(Cu^+) = -1200$, $\eta_\infty(Ag^+) = -8050$, $\eta_\infty(Cs^+) = -670$, $\eta_\infty(Hg^{2+}) = -63\,000$, and $\eta_\infty(In^{3+}) = -3791$.

The purpose of this paper is to report the η_∞ values for the following ions using HFS wave functions: Ti^{3+}, Mn^{2+}, Cu^{+}, Zn^{2+}, Ge^{4+}, Rb^{+}, Zr^{3+}, Nb^{5+}, Tc^{2+}, Ag^{+}, In^{3+}, Sb^{3+}, Cs^{+}, Hf^{3+}, Re^{2+}, Au^{+}, Hg^{2+}, Bi^{3+}, Fr^{+}, Th^{3+}, and Am^{2+}. Although some of these ions, e.g., Cu^{+}, Ag^{+}, Au^{+}, Hg^{2+}, and Th^{3+} do not have nuclear spin $I \geq 2$, their inclusion becomes necessary in order to understand the variation of η_∞ values over the entire region of the Periodic Table. In Sec. II, we briefly outline Sternheimer's theory for the calculation of η_∞ and describe the method of computation adopted in the present work. In Sec. III, we present our results and arrive at a few general conclusions. We also compare our results on Cu^{+}, Ag^{+}, Cs^{+}, In^{3+}, and Hg^{2+} with those obtained by Sternheimer.[12-14]

II. DETAILS OF THE CALCULATIONS

The following derivation and Eqs. (3)–(10) of this section are essentially identical to those of Sternheimer in Refs. 6 and 12. The perturbing potential due to nuclear H, acting on an electron is given by (in Ry)

$$V_H = -HP_4(\theta)/(4r^5), \tag{3}$$

where r is the distance of the electron from the center of the nucleus, P_4 is the fourth-degree Legendre polynomial, and θ is the angle between the position vector of the electron and the nuclear spin direction. The radial distortion $u'_{1,H}(nl \to l)$ of the closed-shell electron wave function $u'_0(nl)$ is obtained from the solution of the inhomogeneous equation

$$\left(\frac{-d^2}{dr^2}+\frac{l'(l'+1)}{r^2}+V_0-E_0\right)u'_{1,H}(nl\to l') = u'_0(nl)\left(\frac{1}{r^5}-\langle 1/r^5\rangle_{nl}\right), \tag{4}$$

where n denotes the principal quantum number of the outermost d or f orbital, V_0 is the effective potential, E_0 is the unperturbed energy eigenvalue, and $\langle 1/r^5\rangle_{nl}$ is the expectation value of $1/r^5$ over $u'_0(nl)$. In actual calculations, V_0-E_0 is replaced by the local approximation

$$V_0-E_0=\frac{1}{u'_0}\frac{d^2u'_0}{dr^2}-\frac{l(l+1)}{r^2}. \tag{5}$$

The unperturbed radial wave function $u'_0(nl)$ is normalized as

$$\int_0^\infty [u'_0(nl)]^2\,dr=1. \tag{6}$$

The perturbed wave function $u'_{1,H}(nl\to l')$ is orthogonalized according to

$$\int_0^\infty u'_0(nl)u'_{1,H}(nl\to l')\,dr=0. \tag{7}$$

The contribution to the total η_∞ due to the radial excitations $nd\to d$ and $nf\to f$ are respectively given by the following equations:

$$\eta_\infty(nd\to d)=\frac{80}{63}\int_0^\infty u'_0(nd)u'_{1,H}(nd\to d)r^4\,dr$$

and (8)

$$\eta_\infty(nf\to f)=\frac{112}{99}\int_0^\infty u'_0(nf)u'_{1,H}(nf\to f)r^4\,dr,$$

where the numerical coefficients result from the integration over the angular parts of the wave functions and their summation over the magnetic substates.

The HFS wave functions for all the ions were generated over 441-point mesh using a modified Herman-Skillman program[11] adopted for an IBM 7044/1401 computer system at I.I.T., Kanpur. The difference equation corresponding to Eq. (4) was integrated inwards starting from the last mesh point r_1 of the $u'_0(nl)$ and the trial value of $[u'_{1,H}(nl)]_{r=r_1}$ was slowly varied in an iterative way till the solution became well behaved near $r=0$.

For $nd\to d$ perturbation, the boundary condition at $r=0$ is given by

$$[u'_{1,H}(nl)]_{r=0}=\tfrac{1}{6}C, \tag{9}$$

where C is the coefficient of r^3 in the power series expansion of $u'_0(nl)$ near the nucleus. Three to four iterations were sufficient to achieve this boundary condition.

For Cu^+, Ag^+, and Au^+ ions the contributions $\eta_{\infty,\mathrm{ang}}$ due to angular excitations were estimated from the Thomas-Fermi relation[6]

$$(\gamma_{\infty,\mathrm{ang}}/\eta_{\infty,\mathrm{ang}})_{\mathrm{HFS}}=\tfrac{9}{5}, \tag{10}$$

where $\gamma_{\infty,\mathrm{ang}}$ is the total angular contribution to the quadrupole antishielding factor,[5] γ_∞, obtained by using HFS wave functions. Because of the negligible magnitude ($\sim+1$) of $\eta_{\infty,\mathrm{ang}}$ as compared to $\eta_{\infty,\mathrm{rad}}$ (-10^3 to -10^5), no attempt was made to calculate $\eta_{\infty,\mathrm{ang}}$ for the other ions and it will be assumed that $\eta_\infty=\eta_{\infty,\mathrm{rad}}$.

All the integrals over the orbitals were evaluated by the method of finite differences[15] using the formula of integration through adjacent intervals with differences up to fourth order included. In cases of the ions with incomplete outermost d or f orbitals the constants in Eq. (8) were multiplied by the fraction to which these shells are filled. The η_∞ values reported here have an estimated error of 10% due to the method of numerical integration of Eq. (4).

III. RESULTS AND DISCUSSION

In Table I we present the values of various individual radial contributions to η_∞ along with the $\langle 1/r^5\rangle$ values for the outermost $u'_0(nd)$ wave function for all the ions studied here. In Fig. 1 we give the plot of the perturbed wave function $u'_{1,H}(5d\to d)$ along with the unperturbed wave function $u'_0(5d)$ (times 2500) for Au^+ ion. The strong antishielding effect arises due to the relatively large magnitude and opposite sign of the perturbed wave function with respect to the unperturbed wave function in the more important region of large r.

In all the cases considered by us the strongest contribution comes from the outermost nd shell. The $nf\to f$ excitations produce a much weaker antishielding as compared to the $nd\to d$ excitations. In Fig. 2 we represent the variation of $|\eta_\infty|$ as a function of the atomic number Z, for the $3d$, $4d$, and $5d$ series of ions, respectively. For all the nd shells it is clear from Fig. 2 that $|\eta_\infty|$ value increases very rapidly as Z increases and reaches a maximum value, respectively, at Cu^+, Ag^+, and Au^+ ions. For these three ions the corresponding nd shell is complete and most external. With further increase in Z, $|\eta_\infty|$ again decreases. Sternheimer[14] earlier observed a similar trend in case

TABLE I. Individual radial contributions to the total nuclear hexadecapole antishielding factor η_∞. $\langle 1/r^5 \rangle_{nd}$ gives the expectation value of $1/r^5$ over the outermost nd orbital.

Ion \ Perturbation	$3d \to d$	$4d \to d$	$4f \to f$	$5d \to d$	$5f \to f$	$6d \to d$	$\langle 1/r^5 \rangle_{nd}$	$\eta_{\infty,\mathrm{rad}}$
Ti^{3+}	−26.234						44.458	−26.234
Mn^{2+}	−274.63						101.13	−274.63
Cu^{+}	−1471.7						262.76	−1471.74
Zn^{2+}	−352.51						361.38	−352.51
Ge^{4+}	−161.58						637.85	−161.58
Rb^{+}	−57.688						1943.1	−57.688
Zr^{3+}	−35.668	−711.85					308.36	−747.53
Nb^{5+}	−30.865						4085.20	−30.865
Tc^{2+}	−25.560	−3947.5					563.04	−3973.1
Ag^{+}	−18.194	−9562.3					1138.2	−9580.4
In^{3+}	−15.757	−2859.1					1841.3	−2875.0
Sb^{3+}	−13.835	−1686.1					2724.2	−1699.9
Cs^{+}	−11.018	−866.58					5319.1	−877.60
Hf^{3+}	−5.833	−376.69	−70.031	−6794.8			2504.2	−7247.4
Re^{2+}	−5.376	−309.18	−33.262	−34 841			3766.7	−35 189
Au^{+}	−4.859	−242.77	−17.010	−71 279			6208.1	−71 544.
Hg^{2+}	−4.743	−229.37	−14.762	−37 141			7606.8	−37 390
Bi^{3+}	−4.423	−194.90	−10.297	−14 506			12 372	−14 716
Fr^{+}	−4.052	−160.165	−7.116	−7818.1			20 687	−7989.4
Th^{3+}	−3.808	−139.98	−5.674	−4956.0		−28 452.	3860.0	−33 557.
Am^{2+}	−3.458	−115.49	−4.270	−4067.2	−803.674		44 280	−4994.1

of $4d$ ions. We note here that the magnitude of ionic charge very significantly changes the value of η_∞ for the isoelectronic cases, e.g., Cu^{+}, Zn^{2+}, and Ge^{4+} in the $3d^{10}$ case, Ag^{+} and In^{3+} in the $4d^{10}$ case and Au^{+} and Hg^{2+} in the $5d^{10}$ case. In Fig. 3 we have plotted the $|\eta_\infty|$ values as a function of Z for $3d^1$, $4d^1$, $5d^1$, along with $3d^5$, $4d^5$, $5d^5$, and $3d^{10}$, $4d^{10}$, $5d^{10}$ cases. As expected, for a given number of nd electrons, $|\eta_\infty|$ increases very rapidly with increase in the value of the principal quantum number n.

The angular contributions to η_∞ in the case of Cu^{+}, Ag^{+}, and Au^{+} ions were estimated using Eq. (10). Our $\gamma_{\infty,\mathrm{ang}}$ results[16] for these ions using HFS wave functions are +1.06, +1.42, and +2.06 giving the total $\eta_{\infty,\mathrm{ang}}$ values as +0.58, +0.79, and +1.15,

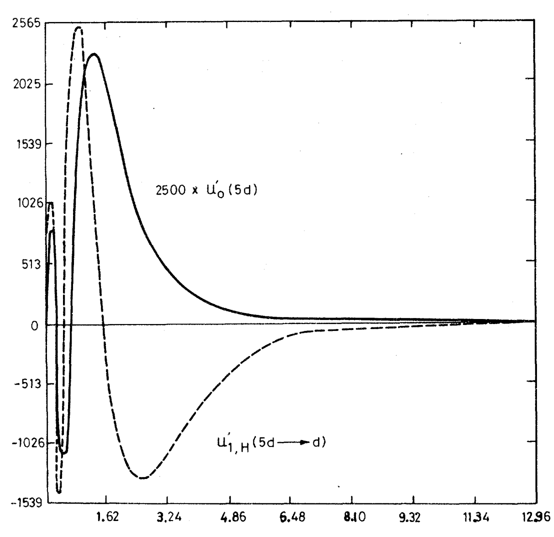

FIG. 1. Perturbed wave function $u'_{1,H}(5d \to d)$ and 2500 times the unperturbed $5d$ function $u_0(5d)$ for Au^{+}.

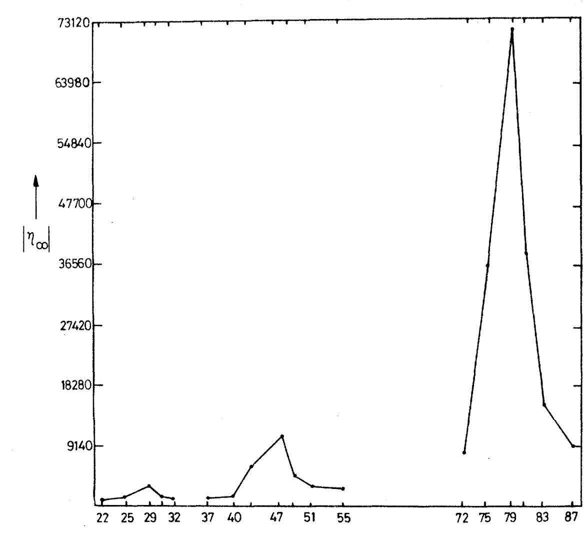

FIG. 2. Variations in $|\eta_\infty|$ values for $3d$, $4d$, and $5d$ ions, respectively, with the atomic number Z.

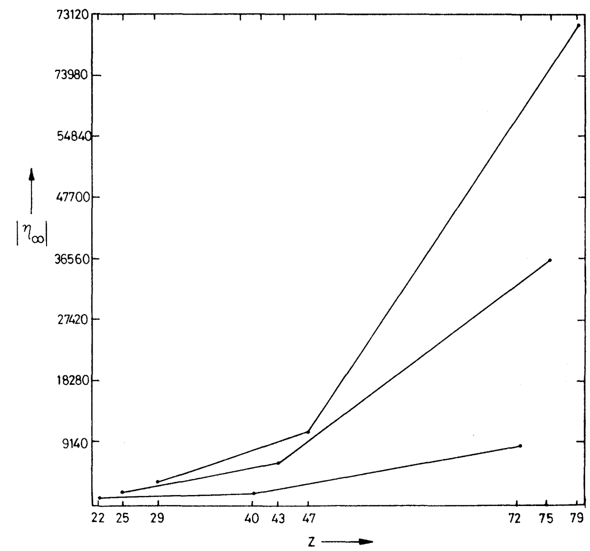

FIG. 3. Variations in $|\eta_\infty|$ values for $3d^1$, $4d^1$, $5d^1$, along with $3d^5$, $4d^5$, $5d^5$, and $3d^{10}$, $4d^{10}$, and $5d^{10}$ ionic cases.

respectively.

It is important to note that the present η_∞ results for Cu^+, Ag^+, and Cs^+ ions are ~1.2 times higher in magnitude than those reported by Sternheimer[12,13] using HF wave functions. The Slater exchange approximation[17] is known to overemphasize the role of exchange. The present calculations on the ions show that at least the outer orbitals are not overcontracted due to this. Saxena and Narasimhan[18] have earlier arrived at similar conclusions from their HFS calculations on the diamagnetic susceptibility and nuclear magnetic shielding for closed shell atoms and ions.

Our test run of η_∞ program using the neutral-atom 441-point-mesh HFS wave functions for In gave $\eta_\infty = -3790$ as compared to Sternheimer's result of -3791. Using ionic wave functions for In^{3+} we obtained η_∞ as -2875. This reduction is due to a significant contraction of the outer $4d$ orbital in In^{3+} ion as compared to the neutral atom.

There have been very few experimental measurements of nuclear hexadecapole moments reported in literature for the ground states of nuclei. It can be anticipated that the nuclei with a large value of the quadrupole moment (Q) and nuclear spin $I \geq 2$ would possess a large hexadecapole moment. In the two regions of the Periodic Table starting from ^{151}Pm ($Z = 61$) to ^{191}Ir ($Z = 77$), and from ^{227}Ac ($Z = 89$) to ^{243}Am ($Z = 95$) the nuclei possess a large quadrupole moment enhanced because of collective effects. Extensive evidence has been obtained for the existence of hexadecapole deformation in the nuclei of rare-earth[19-21] ($A = 152-178$) and actinide[22] ($A = 230-248$) regions and several tungsten nuclei[23] ($A = 179, 181, 183$, and 185). Nuclei in these two regions ($Z = 61-77$ and $Z = 89-95$) with spin $I \geq 2$ would be good candidates for observing nuclear hexadecapole interaction if the corresponding values of η_∞ are also large. Systems in these regions for which large $|\eta_\infty|$ values have been obtained in the present calculations are Au^+, Hg^{2+}, Re^{2+}, Th^{3+}, Bi^{3+}, Fr^+, Hf^{3+}, and Am^{2+}. Of these, nuclei of Au^+ and Hg^{2+} do not satisfy the $I \geq 2$ condition. Naturally abundant ^{232}Th does not have a spin. It appears to us that in this region Re^{2+} and Bi^{3+} are most likely to exhibit hexadecapole coupling effects judging from their Q value, spin, natural abundance, magnetic resonance sensitivity, and η_∞ values. ^{55}Mn, although it does not fall in the high-Z region, may also be mentioned here on account of its spin, natural abundance, and resonance sensitivity in comparison to a few other nuclei considered here. We have added the magnetic-resonance sensitivity consideration on account of the experimental aspect of detecting the hexadecapole interaction via magnetic resonance.

In Table II we present the results of our calculations of η_∞ for Nb^+, Nb^{2+}, Nb^{3+}, Nb^{4+}, and Nb^{5+} cases. Niobium satisfies spin, quadrupole moment, abundance and sensitivity criteria. In covalent compounds of niobium it is likely that one gets contributions to η_∞ from both $3d^{10}$ and $4d^n$ orbitals. It is clear from Table II that in such cases the effective $|\eta_\infty|$ might also become large. It is there-

TABLE II. Individual radial contributions to the total nuclear hexadecapole antishielding factor η_∞ for Nb^+, Nb^{2+}, Nb^{3+}, Nb^{4+}, and Nb^{5+} ions. $\langle 1/r^5 \rangle_{nd}$ gives the expectation value of $1/r^5$ over the outermost nd orbital.

Ion \ Perturbation	$3d \to d$	$4d \to d$	$\langle 1/r^5 \rangle_{nd}$	$\eta_{\infty,\mathrm{rad}}$
Nb^+	−31.813	−11 039.1	299.05	−11 071.0
Nb^{2+}	−31.716	−3300.8	348.29	−3332.5
Nb^{3+}	−31.531	−1209.8	399.07	−1241.3
Nb^{4+}	−31.249	−390.52	451.11	−421.77
Nb^{5+}	−30.865		4085.2	−30.865

fore not surprising that the observation of hexadecapole coupling for this nucleus has been reported[3] in $NbCl_5$. In the case of indium and antimony the $4d^{10}$ configuration gives rise to high-$|\eta_\infty|$ values and since these nuclei also satisfy the other criteria mentioned above we can expect to observe hexadecapole coupling effects here. Indeed such effects have been observed[1,2] in the cases of antimony in $SbCl_5$ and indium in InAs. It should be pointed out here that while the detection of hexadecapole couplings via nuclear quadrupole coupling effects is facilitated by high $|\eta_\infty|$ and $\partial^3 E_z/\partial z_i^3 \neq 0$, it also requires the presence of unfilled p shells around the nucleus in question since only then can we hope to observe quadrupolar effects. The recent observation[4] of hexadecapole coupling in the ground state of ^{165}Ho using the atomic-beam magnetic resonance method is highly encouraging and appears to be a promising way of obtaining information regarding nuclear hexadecapole moments.

For ^{185}Re and ^{187}Re ($Q \approx 2.66$ b) Sternheimer[13] has suggested that η_∞ value might be very large, perhaps of the same order as that of Hg^{2+}. Our results, −34 841 and −37 141, respectively, for Re^{2+} and Hg^{2+}, show that this is indeed true. A higher $|\eta_\infty|$ value in the case of Hg^{2+} suggests that the number of $5d$ electrons being twice that in Re^{2+} more than compensates for the effect due to a tighter binding of these electrons. As compared to Sternheimer's result of $\eta_\infty(Hg^{2+}) = -63\,000$, our $|\eta_\infty|$ result is approximately a factor of 2 smaller. This is due to the fact that Hartree wave functions have been used by Sternheimer and these do not include exchange and therefore are more external than the HFS wave functions. This also clearly brings out the sensitivity of the η_∞ result to the quality of the wave function.

Coming now to the heaviest atom considered in the present calculations, namely Am, we see that for ^{241}Am and ^{243}Am ($Q = 4.9$ b) the contribution to η_∞ from the half-filled $5f$ shell is quite appreciable (−803.67 as compared to −4.27 from the completely filled $4f$ shell). Also as compared to the other $5d$ ions we considered, the $5d$ orbital in Am^{2+} is much more internal which makes $\eta_\infty(Am^{2+}) = -4994.16$.

It is surprising to note the strong antishielding ($\eta_\infty = -28\,542$) produced due to $6d \to d$ excitation in Th^{3+} ($Z = 90$) ion which has a single electron in the $6d$ orbital. This suggests that the nuclei around the region of $Z = 90$ with spin $I \geq 2$ are very good candidates for detecting electric hexadecapole interaction. However, from the experimental point of view the problem of abundance becomes quite serious in this region.

IV. SUMMARY

In the present work we have calculated η_∞ for 21 ions listed in Sec. I. A majority of them can possess nuclear hexadecapole moment. For nd shell ions, with $n = 3, 4$ and 5 respectively, it is pointed out that the maximum $|\eta_\infty|$ value is obtained where the outermost d shell is just complete. With a change in Z in either direction the $|\eta_\infty|$ decreases very rapidly for a given value of the principal quantum number n of the outermost d shell. Although the excitations $nf \to f$ produce a much weaker antishielding effect than the $nd \to d$ excitations in the case of Am^{2+}, the contribution from half-filled $5f$ shell is quite significant. The use of ionic wave functions for the unperturbed function leads to a significant reduction in $|\eta_\infty|$ values, e.g., from 3790 for neutral In to 2875 for In^{3+} ion. The very large $|\eta_\infty|$ obtained in the case of Re^{2+}, and Bi^{3+} along with the favorable parameters such as spin, natural abundance, and magnetic resonance sensitivity possessed by them suggests that these ions are, besides the already investigated cases of In^{3+} and Sb^{5+}, very good candidates for experimental detection of nuclear hexadecapole interaction. The case of Mn^{2+} also appears to be prominent in this regard.

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